



## Synthesis of Chitosan/Graphene Oxide-Ferric Oxide Composite via Coprecipitation: An Upgraded Adsorbent Material for Methylene Blue Dye Removal

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### Abstract:

This study addresses the critical need for effective dye removal in wastewater treatment by developing a chitosan/graphene oxide-ferric oxide (CS/GO-Fe<sub>2</sub>O<sub>3</sub>) composite through a coprecipitation method. The primary objective was to evaluate its adsorption performance for methylene blue (MB), a model cationic dye, while optimizing key operational parameters. The composite was systematically characterized using X-ray diffraction (XRD), scanning electron microscopy-energy dispersive X-ray spectroscopy (SEM-EDS), and Brunauer-Emmett-Teller (BET) analysis. XRD patterns confirmed the integration of crystalline Fe<sub>2</sub>O<sub>3</sub> nanoparticles with a crystallite size of 19.25 nm within the composite matrix, while SEM-EDS revealed a transformation from a dense chitosan morphology to a highly porous structure in the CS/GO-Fe<sub>2</sub>O<sub>3</sub> composite enriched with carbon, oxygen, nitrogen, and iron. BET analysis demonstrated a substantially higher surface area than that of conventional adsorbents, increasing from 7.036 m<sup>2</sup>/g for pure chitosan to 39.946 m<sup>2</sup>/g for the composite. Adsorption studies identified the optimal conditions at pH 9 and a contact time of 50 minutes. The composite exhibited superior adsorption capacity relative to many reported materials, following the Langmuir isotherm model, which has a maximum adsorption capacity (Q<sub>m</sub>) of 11.655 mg/g. These findings highlight the composite's potential as a cost-effective and magnetically separable adsorbent for environmental remediation, offering insights into the design of hybrid materials for organic pollutant removal.

### Keywords:

Adsorption dye; Chitosan; Ferric oxide; Graphene oxide; Methylene blue.

### Highlights:

- The chitosan/graphene oxide-ferric oxide composite shows composite has a porous structure with a large surface area (39.946 m<sup>2</sup>/g).
- The ability of chitosan/ chitosan/graphene oxide-ferric oxide composite to adsorb methylene blue dye has a very high adsorption capacity and adsorption efficiency.
- The adsorption isotherm model follows the Langmuir model.

### Article History:

|                           |              |
|---------------------------|--------------|
| Received:                 | 22 Feb. 2025 |
| Received in revised form: | 02 May 2025  |
| Accepted:                 | 06 Jan. 2026 |
| Final Proofreading:       | 09 May 2026  |
| Available online:         | 15 Sep. 2026 |

### Citation:

Desnelli, Syafutri F, Alfarado D, Musifa E, Said M, Mara A, Suheryanto, Andriyanti W, Kartika BM, Wahyudiyanto H. **Synthesis of Chitosan/Graphene Oxide-Ferric Oxide Composite via Coprecipitation: An Upgraded Adsorbent Material for Methylene Blue Dye Removal.** *Tikrit Journal of Engineering Sciences* 2026; 33(2): 2502.

<https://doi.org/10.25130/tjes.33.2.3>

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## 1. INTRODUCTION

Dye pollution remains a critical environmental challenge, with over 700,000 tons of synthetic dyes produced annually worldwide, of which 10–15% are discharged untreated into water systems [1]. Dyes are organic compounds that exhibit resistance and toxicity. One widely used dye in industry is methylene blue [2]. Methylene blue is a type of blue, cationic, and thiazine dye widely applied in the textile industry as a fiber dye [3]. Methylene blue has harmful effects on human health and ecosystems, so removing it from wastewater is necessary because it increases toxicity in water systems [4]. Several methods exist to address this dye problem, including electrocoagulation, oxidation, photocatalysis, degradation, biodegradation, biocatalytic degradation, and adsorption [5]. Conventional wastewater treatments such as electrocoagulation and photocatalysis often face limitations in efficiency, cost, and scalability, whereas adsorption has emerged as a superior approach due to its operational simplicity, cost-effectiveness, and adaptability to diverse pollutants [6–8]. Commonly known adsorbents include activated carbon, molecular sieves, and polymer adsorbents [9]. One widely used adsorbent is chitosan. Chitosan is used to remove dyes obtained from natural resources. Chitosan is produced by the deacetylation of chitin and has a lower storage capacity than cellulose. Chitosan has several advantages compared to other commercial adsorbents, which offer a sustainable alternative with intrinsic advantages: biodegradability, abundant hydroxyl/amino groups for pollutant binding, and low toxicity [10]. Chitosan, as an adsorbent, is a naturally biodegradable polymer material with many hydroxyl groups and highly reactive amino groups [11]. However, raw chitosan exhibits poor mechanical stability in acidic conditions and moderate adsorption capacity [12]. Chitosan can be combined with graphene oxide and  $\text{Fe}_2\text{O}_3$  into a composite for liquid waste treatment. Recent advancements in composite materials have addressed these limitations by integrating chitosan with carbon nanomaterials, such as graphene oxide (GO). Graphene is an auspicious carbon material and has attracted significant attention as an adsorbent due to its high adsorption capacity. Graphene and Graphene Oxide (GO) have good chemical stability. Graphene-based composites can be used to adsorb various pollutants from water, including heavy metals, pesticides, antibiotics, and dyes [13]. Graphene oxide has a large surface area and is rich in oxygen functional groups that can bind to metal ions. GO's high surface area (theoretical  $\sim 2630 \text{ m}^2/\text{g}$ ) and oxygen-rich functional groups (e.g., epoxy, carboxyl) enhance pollutant binding via  $\pi$ - $\pi$  interactions and electrostatic forces [14]. GO/chitosan composites demonstrated a 35%

increase in MB adsorption over pure chitosan, achieving a  $\sim 350 \text{ mg/g}$  capacity [15]. Nevertheless, GO's tendency to aggregate in aqueous media and challenges in post-adsorption recovery remain unresolved issues. To overcome these drawbacks, researchers have incorporated magnetic components such as  $\text{Fe}_3\text{O}_4$  or  $\text{Fe}_2\text{O}_3$  into composites [16]. These materials enable rapid separation via external magnets, reducing secondary pollution risks.  $\text{Fe}_2\text{O}_3$  inorganic compound named Iron (III) Oxide.  $\text{Fe}_2\text{O}_3$  is one type of metal oxide that can be used as an adsorbent due to the electrostatic interaction between the adsorbent and the dye.  $\text{Fe}_2\text{O}_3$  is added to the chitosan/GO composite because it exhibits strong adsorption properties toward various contaminants, thereby increasing the adsorption efficiency of pollutants [17].  $\text{Fe}_2\text{O}_3$  can also support and strengthen the composite structure, providing additional stability and increasing adsorption capacity through surface interactions and active sites. Research by Zhong et al [18] stated that  $\beta$ -cyclodextrin-modified magnetic GO achieved  $89.15 \text{ mg/g}$  Cr(VI) adsorption with 85% retention after five cycles. Similarly,  $\text{Fe}_2\text{O}_3$ -enhanced composites exhibit improved structural integrity and active-site accessibility, as evidenced by clay/GO/ $\text{Fe}_2\text{O}_3$  nanocomposites showing 95% MB removal within 50 minutes [19]. This makes  $\text{Fe}_2\text{O}_3$  a potential material for dye waste processing applications [20]. Despite these advances, gaps persist in optimizing the synergistic effects among chitosan, GO, and  $\text{Fe}_2\text{O}_3$ , particularly in tailoring pore structures and surface charge for cationic dyes such as MB. This study introduces a chitosan/GO-  $\text{Fe}_2\text{O}_3$  composite synthesized via coprecipitation, a method chosen for its simplicity, low energy input, and scalability [21, 22]. The composite leverages chitosan's functional groups, GO's high surface area, and  $\text{Fe}_2\text{O}_3$  magnetic responsiveness to address three critical limitations of prior studies: Aggregation Mitigation ( $\text{Fe}_2\text{O}_3$  nanoparticles act as spacers between GO sheets, reducing restacking and exposing more binding sites) [23, 24]; acid stability material (crosslinking chitosan with GO enhances structural resilience in acidic conditions, a weakness of pure chitosan) [25,26]; and for recyclability (magnetic separation eliminates the need for energy-intensive filtration, addressing GO's recovery challenges) [27]. This work advances the design of multifunctional adsorbents by elucidating structure-property relationships and providing a sustainable solution for the remediation of industrial dye wastewater.

## 2. MATERIALS AND METHODS

### 2.1. Materials and Instrumentation

The tools that used in this study included a collection of glassware, dropper pipettes, volume pipettes, Whatman No. 42 filter paper,

Shaker, magnetic stirrer, analytical balance, hot plate, oven, pH meter, vacuum pump, stirring rod, spatula, water bath, ice bath, ultrasonication, UV-Vis spectrophotometer Orion Aqua mate 8000, XRD type X'pert PRO, SEM-EDS Thermo Fisher Scientific Phenom P, Series, and BET SSA Full Isothermal 20pts instrument. The materials that used in this study included distilled water, distilled water, methylene blue Merck, chitosan, potassium permanganate ( $\text{KMnO}_4$ ) Merck, sodium nitrate ( $\text{NaNO}_3$ ) Merck, sodium hydroxide ( $\text{NaOH}$ ) Merck, sulphuric acid 96% ( $\text{H}_2\text{SO}_4$ ) SMART-LAB, ammonium hydroxide 25% ( $\text{NH}_4\text{OH}$ ) Merck, iron (III) chloride ( $\text{FeCl}_3$ ) Merck, graphite powder Merck, hydrogen peroxide 30% ( $\text{H}_2\text{O}_2$ ) Merck, glutaraldehyde-methanol technically, ethanol ( $\text{C}_2\text{H}_6\text{O}$ ) Merck, acetic acid ( $\text{CH}_3\text{COOH}$ ) Merck, iron (III) chloride ( $\text{FeCl}_3$ ) Merck, and iron (II) chloride ( $\text{FeCl}_2$ ) Merck.

## 2.2.Synthesis Graphene Oxide

1 gram of graphite powder was oxidized using 50 mL of 96% sulphuric acid, stirred for 24 hours, then 3 grams of sodium nitrate were added and stirred with a water bath containing ice cubes for 1 hour, 6 grams of potassium permanganate were added gradually, then stirred for 4 hours. 30% hydrogen peroxide solution was added to a 50 mL measuring flask to a final volume of 6 mL. After that, it was stirred for 1 hour, and 500 mL of distilled water was added. The solution was then washed until the pH reached 7, and after washing, it was ultrasonicated for 2 hours. The filtered solution was used with filter paper, and the precipitate was dried in an oven at 150 °C for 70 minutes. So that graphene oxide solids were obtained. The experimental schematic is shown in Fig. 1.

## 2.3.Synthesis $\text{Fe}_2\text{O}_3$ Nanoparticles

The  $\text{Fe}_2\text{O}_3$  nanoparticles were synthesized using the coprecipitation method. The  $\text{Fe}^{3+}$  precursor solution was prepared by dissolving 1.4625 g of iron (III) chloride in a 100 mL beaker using distilled water. Next, 12% ammonium hydroxide was added to the solution and stirred continuously until precipitation formed, gradually raising the pH to 10. The resulting precipitate was washed and filtered with distilled water until the pH was neutral, then oven-dried at 150°C for 70 minutes. The  $\text{Fe}_2\text{O}_3$  precipitate was ground and calcined at 300°C for 4 hours, and after synthesis, the composite was characterized using XRD. The experimental schematic is shown in Fig. 1.

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## 2.4.Preparation of Chitosan/GO Composite

The synthesized chitosan/GO composite was prepared using a 5:1 chitosan:graphene oxide ratio. Graphene oxide (GO) 0.8 g was added to 200 mL of a 1.5% acetic acid solution and stirred ultrasonically for 1 hour to ensure complete mixing. 4.0 g of chitosan was dissolved in this solution, stirred ultrasonically to disperse, and then stored in a dark environment for 12 hours. After that, this mixture was dripped into a sodium hydroxide 5% solution at a rate of about one drop per second and left for 24 hours until a composite was formed. The composite was washed with deionized water until the solution pH was around 7.0, then placed in a 5% glutaraldehyde-methanol solution and stirred for 2 hours. Then, it was washed with 50 mL ethanol and 50 mL deionized water respectively several times and dried at 60 °C. The experimental schematic is shown in Fig. 1.

## 2.5.Preparation of Chitosan/GO- $\text{Fe}_2\text{O}_3$ Composite

The synthesized chitosan/GO- $\text{Fe}_2\text{O}_3$  composite was prepared using a Chitosan/GO:  $\text{FeCl}_3$  1:1.6 ratio. 1 gram of chitosan/GO was mixed with 100 mL iron (III) chloride 100 mM solution. The solution was evaporated at 100 °C and then washed with deionized water. The product obtained was dried at 60 °C. The chitosan/GO- $\text{Fe}_2\text{O}_3$  composite obtained was characterized using XRD, SEM-EDS, and BET Electrodes. The experimental schematic is shown in Fig. 1.

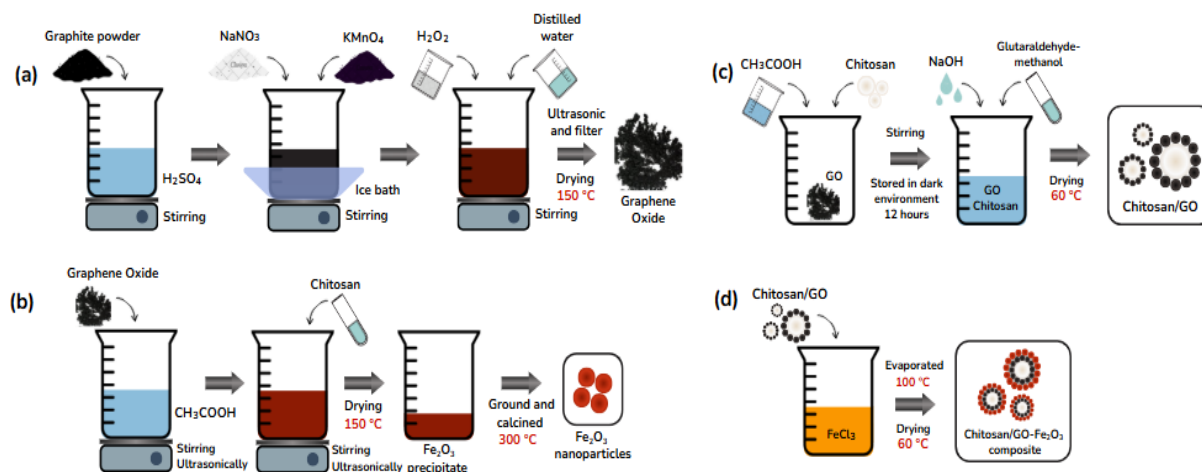


**Fig. 1** Schematic of the Synthesis Procedure of (a) Graphene oxide, (b)  $\text{Fe}_2\text{O}_3$ , (c) Chitosan/GO, and (d) Chitosan/GO- $\text{Fe}_2\text{O}_3$

## 2.6. Study Adsorption of Methylene Blue

### 2.6.1. Calibration Curve for Methylene Blue Standard Solution

A series of standard solutions, 1, 2, 5, 10, 15, 20, 25, 30 ppm, was measured in triplicate to ensure reproducibility. The quantitative analysis of methylene blue (MB) concentration was conducted using UV-Vis spectroscopy with absorbance measurements taken at the characteristic maximum wavelength for  $\lambda_{\max} = 664$  nm. A calibration curve was constructed by plotting the average absorbance against the corresponding known concentration. The relationship exhibited excellent linearity within the experimental concentration range, yielding a calibration equation of with a correlation coefficient ( $R^2$ ). The high  $R^2$  value confirms a strong linear relationship, verifying the method's adherence to the Beer-Lambert law within our working range. Furthermore, the blank (deionized water) and control samples were routinely measured to account for any baseline drift or instrumental error. The concentration of methylene blue in the unknown experimental samples was then calculated directly using this validated calibration equation.

### 2.6.2. Adsorption of Methylene Blue Solution Based on the Effect of pH

Methylene blue solution 15 mL (25 ppm) was put into a beaker. Then, the pH was adjusted

with pH variations (7, 8, 9, 10, and 11) by adding 0.01 M hydrogen chloride and 0.01 M sodium hydroxide. Next, 0.05 g of chitosan/GO- $\text{Fe}_2\text{O}_3$  composite was added to each adjusted pH. The mixture was stirred for 50 minutes. Afterward, it was analyzed using a visible-light spectrophotometer at 664 nm.

### 2.6.3. Adsorption of Methylene Blue Solution Based on the Effect of Contact Time

A total of 15 mL of a 20 ppm methylene blue solution was placed in an Erlenmeyer flask, and 0.05 g of the chitosan/GO- $\text{Fe}_2\text{O}_3$  composite was added. Then, the mixture was stirred with contact time variations (10, 20, 30, 40, 50, and 60) minutes. Afterward, it was analyzed using a visible-light spectrophotometer at 664 nm. The experimental schematic is shown in Fig. 2.

### 2.6.4. Adsorption of Methylene Blue Solution Based on the Effect of Concentration

A total of 15 mL of methylene blue solution at concentrations of 25, 30, 35, 40, and 45 ppm was placed in an Erlenmeyer flask, and 0.05 g of chitosan/GO- $\text{Fe}_2\text{O}_3$  composite was added. The mixture was stirred at the optimum contact time. After that, it was analyzed using a visible-light spectrophotometer at 664 nm. The experimental schematic is shown in Fig. 2.

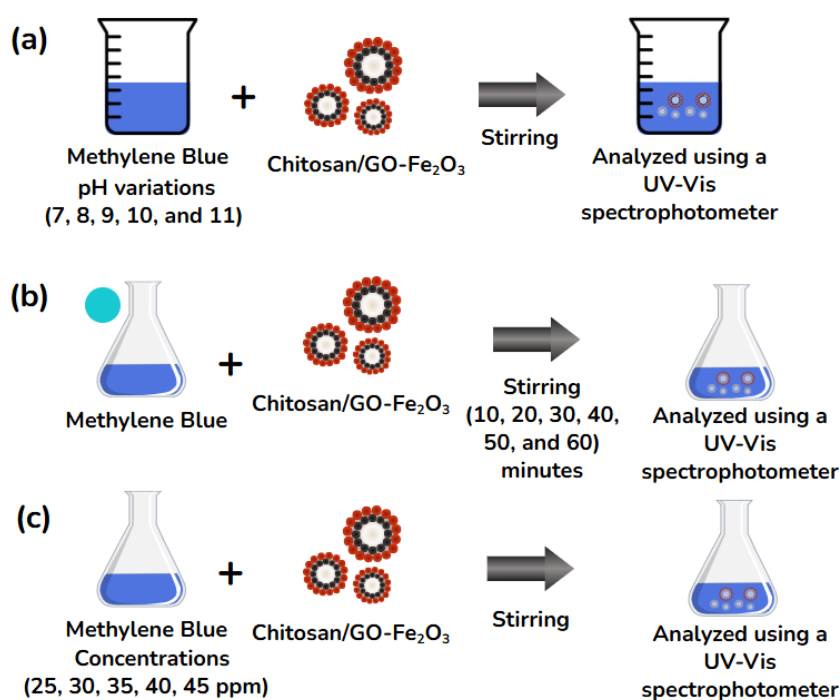


**Fig. 2** (a) Effect of pH, (b) Effect of Contact Time, and (c) Effect of Concentration.

## 2.7. Data Analysis

The results of synthesizing the chitosan/GO- $\text{Fe}_2\text{O}_3$  composite were characterized using XRD, SEM-EDS, and BET instruments. XRD characterization aims to determine the crystal size of the synthesized composite. The XRD analysis will produce a diffractogram. The spectrum of the resulting sample will be

compared with the spectrum pattern contained in the Joint Committee on Powder Diffraction Standards (JCPDS). The diffraction pattern of XRD is plotted as a function of  $2\theta$ , where  $\theta$  represents the diffraction angle. The crystallite size of the chitosan/GO- $\text{Fe}_2\text{O}_3$  composite can be calculated using the following Debye Scherrer Eq. (1):

$$d = \frac{k\lambda}{\beta \cos \theta} \quad (1)$$

Furthermore, the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite was characterized using the SEM-EDS instrument. SEM analysis aims to determine the surface morphology of the synthesized composite. In contrast, EDS analysis will provide data as percentages of the composition of the test sample. The characterized composite will be applied to adsorb the methylene blue dye. After the methylene blue solution adsorption, the absorbance value will be measured using a UV-visible spectrophotometer. The concentration of methylene blue after adsorption can be determined by entering the absorbance value into the following linear regression Eq. (2):

$$y = ax + b \quad (2)$$

Where y represents absorbance, a represents slope, x represents residual concentration, and b represents intercept. After obtaining the residual concentration of methylene blue solution, the percentage of concentration reduction can be calculated using the following Eq. (3):

$$P = \frac{(C_0 - C_t)}{C_0} \times 100\% \quad (3)$$

The general adsorption isotherm models commonly used are the Langmuir and Freundlich isotherms, which determine the adsorption isotherm model on the adsorbent. The Langmuir adsorption isotherm model equation is Eq. (4):

$$Q = \frac{b, K_L, C_e}{1 + K_L, C_e} \quad (4)$$

The equation above can be derived linearly, including:

$$\frac{C_e}{Q} = \frac{1}{K_L} + \frac{1}{b} C_e \quad (5)$$

The Freundlich adsorption isotherm Eq. (6) is:

$$Q = K_F \cdot C_e^{1/2} \quad (6)$$

The Eq. (6) above can be derived linearly:

$$\log Q = \log K_F + \frac{1}{n} \log C_e \quad (7)$$

The adsorption capacity is calculated using Eq. (8):

$$Q_e = \frac{(C_0 - C_e) \times V}{W} \quad (8)$$

The percentage decrease concentration of methylene blue can be calculated using Eq. (9) below:

$$\% \text{Adsorption efficiency} = \frac{C_0 - C_e}{C_0} \times 100\% \quad (9)$$

The kinetics of the adsorption rate were determined using pseudo-first order (10) and pseudo-second order (11) kinetic models.

$$\log(q_e - q_t) = \log q_e - \left[ \frac{k_1}{2.303} \right] t \quad (10)$$

$$\frac{t}{q_t} = \frac{1}{k^2 q_e^2} + \frac{1}{q_e} t \quad (11)$$

### 3. RESULTS AND DISCUSSION

#### 3.1. Characterization of Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composite Material

##### 3.1.1. Characterization of Chitosan, Fe<sub>2</sub>O<sub>3</sub>, Graphene Oxide (GO), Chitosan/GO and Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composites Using XRD

Characterization using XRD was performed on chitosan, Fe<sub>2</sub>O<sub>3</sub>, and Graphene Oxide (GO). The results of XRD characterization are shown in Fig. 3 and Fig. 4. Based on Fig. 3(a), the XRD pattern of chitosan shows a peak at  $2\theta = 20.86^\circ$  with an FWHM of 8.8253. This is similar to the research conducted by Liu et al. [28], namely, the chitosan peak is at  $2\theta = 20^\circ$ . The chitosan peak used in this study has an amorphous structure or lacks a regular crystal structure, as indicated by a gentle/wide diffraction peak. Fig. 3(b), the results of the Fe<sub>2</sub>O<sub>3</sub> diffractogram produced peaks of  $2\theta = 24.15^\circ, 33.16^\circ, 35.63^\circ$  and  $54.04^\circ$ . The diffractogram results of Fe<sub>2</sub>O<sub>3</sub> are similar to the JCPDS No. 33-0664 in that Fe<sub>2</sub>O<sub>3</sub> has peaks of  $2\theta = 24.13^\circ, 33.15^\circ, 35.61^\circ$  and  $54.08^\circ$ . Fig. 3(c) results of XRD characterization of have sharp peaks with angles of  $2\theta = 10.94$  and  $42.52$  according to research conducted by Liu et al. [29], where the GO peak is around  $2\theta = 10.71$  and  $42.35$ . This shows that GO was successfully synthesized.

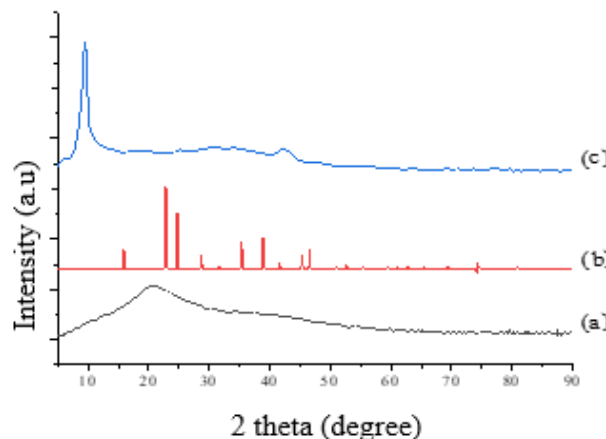


**Fig. 3** XRD Results from: (a) Chitosan, (b) Fe<sub>2</sub>O<sub>3</sub> and (c) Graphene Oxide (GO).

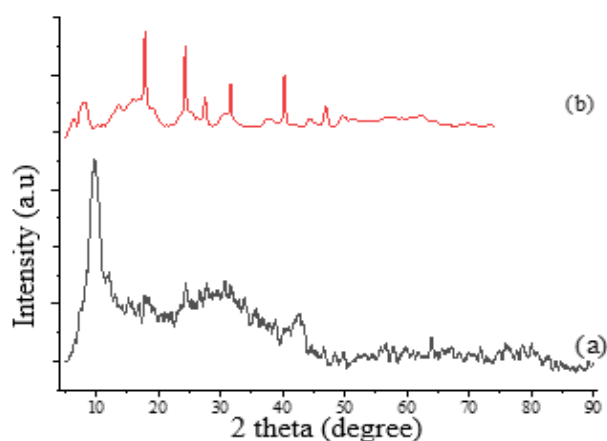


**Fig. 4** XRD Result from: (a) Chitosan/GO, (b) Chitosan/GO Fe<sub>2</sub>O<sub>3</sub>.

Based on Fig. 4(a), the results of the characterization of the chitosan/GO composite obtained chitosan peaks  $2\theta = 18.47^\circ$ ,  $19.22^\circ$ , which are associated with the amorphous state of chitosan and GO peaks  $2\theta = 9.68^\circ$ , which are associated with GO. The sharp GO peaks in the composite experience an angular shift from  $2\theta = 10.71^\circ$  to  $9.68^\circ$ . The chitosan/GO composite shows a more dominant GO peak than the chitosan peak, so the degree of chitosan crystallization decreases after adding GO. In addition, chitosan is evenly distributed in the GO matrix [30]. Figure 4(b), where the diffractogram results of the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite have peaks at angles of  $26.70^\circ$ ,  $35.11^\circ$ ,  $39.25^\circ$ ,  $44.59^\circ$ ,  $55.85^\circ$ , and  $64.49^\circ$ . It can be compared with JCPDS NO.75-0449 data at an angle of  $2\theta$ , namely  $22.52^\circ$ ,  $30.1^\circ$ ,  $35.8^\circ$ ,  $43.5^\circ$ ,  $57.5^\circ$ ,  $63.1^\circ$  and JCBDS No. 33-0664, namely  $24.97^\circ$ ,  $34.23^\circ$ ,  $36.63^\circ$ ,  $50.71^\circ$ , and  $55.17^\circ$  which indicate the presence of rhombohedral  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> crystals [31]. When dispersed in a chitosan/GO matrix, Fe<sub>2</sub>O<sub>3</sub> will occupy space within the polymer matrix. Fe<sub>2</sub>O<sub>3</sub> has a crystalline structure, so when dispersed into a chitosan/GO matrix, the crystalline peaks of Fe<sub>2</sub>O<sub>3</sub> are more dominant, visually and quantitatively dominating the XRD pattern, giving the impression that the composite is more crystalline. However, the diffraction pattern of the chitosan/GO/Fe<sub>2</sub>O<sub>3</sub> composite at the  $2\theta$  peak around 20 degrees has a wide diffraction pattern, which indicates that chitosan has an amorphous structure. The presence of dispersed Fe<sub>2</sub>O<sub>3</sub> disrupts the regularity of the Chitosan chains, which indirectly increases the amorphous fraction of the matrix. The amorphous structure of the polymer in the Chitosan/GO/Fe<sub>2</sub>O<sub>3</sub> composite plays a very important role because it produces high porosity and a large surface area so that it can be used as an adsorbent in adsorbing methylene blue dye. XRD characterization can also be used to determine the crystal size. The crystal size of the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite was calculated using the Debye-Scherrer formula so that the chitosan/GO-

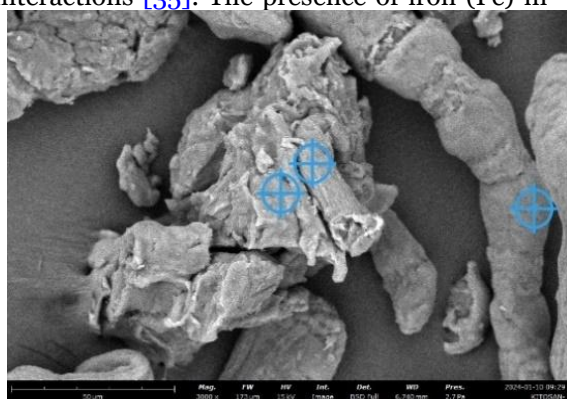
Fe<sub>2</sub>O<sub>3</sub> size was obtained as 19.25 nm. Specifically, the crystallite size of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub>, as determined using the Debye-Scherrer formula, was reported to be approximately 19.25 nm. This is consistent with data from several studies showing that the crystallite size of Fe<sub>2</sub>O<sub>3</sub> in similar composites ranges from 16.5 nm to 22 nm, depending on synthesis method and material composition [32]. The crystallite size of 19.25 nm indicates that the Fe<sub>2</sub>O<sub>3</sub> particles in the chitosan/GO composite are fairly small, which generally provides superior physical and chemical properties, such as a large surface area and good catalytic or adsorption activity. Therefore, the crystallite size of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> of 19.25 nm calculated by the Debye-Scherrer formula is a reasonable value and consistent with the literature related to Fe<sub>2</sub>O<sub>3</sub> and graphene oxide-based nanocomposites reported in various scientific studies [33].

### 3.1.2.Characterization of Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composite Using SEM-EDS

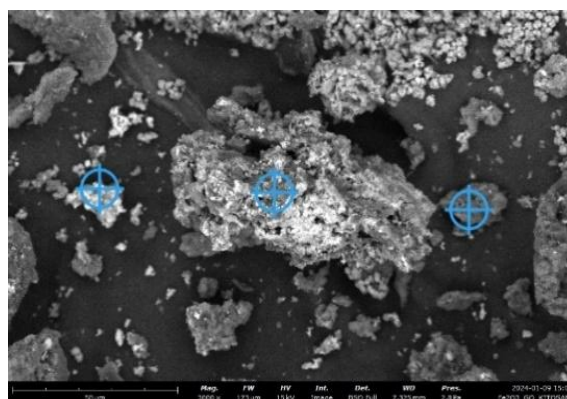
SEM-EDS characterization was performed on chitosan and chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite samples. Characterization using SEM-EDS aims to compare the surface morphology of chitosan and chitosan/GO-Fe<sub>2</sub>O<sub>3</sub>. SEM characterization of chitosan and chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> can be seen in Fig. 5 and Table 1. Based on Fig. 5(a), chitosan has a dense, solid surface. Chitosan has a rhombic crystal form with a cross-linked structure between alpha, beta and gamma forms, forming a resin-like matrix suitable for use as an absorbent or immobilization agent [34]. Figure 5 (b) shows that the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite has a porous structure. According to research, the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite has a rough, rocky surface due to the incorporation of GO into chitosan. After Fe<sub>2</sub>O<sub>3</sub> was added to the GO/chitosan mixture, and the granules were attached to the surface of the adsorbent so that the granules would bond to each other, forming many pores. Because it has pores, the composite can be used as an adsorbent in

adsorbing methylene blue dye. The EDS test determines the elements present in chitosan and chitosan/GO-Fe<sub>2</sub>O<sub>3</sub>. The results of the EDS characterization was shown in Table 1. Based on the results of Energy Dispersive Spectroscopy (EDS) analysis, the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite shows the content of major elements such as carbon (C), nitrogen (N), oxygen (O), sodium (Na), sulfur (S), chlorine (Cl), potassium (K), manganese (Mn), and iron (Fe). The increase in carbon content from 43.04% in pure chitosan to 54% in the composite indicates the successful integration of carbon-rich graphene oxide (GO) into the chitosan matrix. GO has a two-dimensional sheet structure with oxygen-containing functional groups, such as epoxide, hydroxyl, and carboxyl, that can interact chemically with the amine groups of chitosan, forming a homogeneous and stable composite network. The decrease in oxygen content in the composite indicates partial reduction of GO during synthesis, which increases conductivity and intercomponent interactions [35]. The presence of iron (Fe) in

the composite confirms the incorporation of Fe<sub>2</sub>O<sub>3</sub> nanoparticles, which are known to exhibit magnetic properties and produce reactive oxygen species (ROS) that can enhance the composite's antibacterial and catalytic activities [36]. In addition, nitrogen (N) originating from the amine group of chitosan was still detected, indicating that the chemical structure of chitosan was maintained and acted as an active site in the adsorption of metal ions and dyes [37]. The pore structure formed on the surface of the composite, observed by SEM, also contributed to an increase in specific surface area, thereby enhancing the composite's adsorption capacity for pollutants such as methylene blue. Overall, significant changes in elemental composition - increased carbon, decreased oxygen, and the presence of iron and nitrogen - confirmed the successful synthesis of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composites that have an integrated structure and improved functional properties [36]



(a)



(b)

**Fig. 5** SEM Morphology 3000x Resolution of: (a) Chitosan and (b) Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub>.

**Table 1** Composition of Constituent Elements Found in Chitosan and Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Based on EDS Results.

| Material                                    | Elements (% mass) |     |      | Na  | S   | Cl  | K   | Mn  | Fe |
|---------------------------------------------|-------------------|-----|------|-----|-----|-----|-----|-----|----|
|                                             | C                 | N   | O    |     |     |     |     |     |    |
| Chitosan                                    | 43                | 3.9 | 54   | -   | -   | -   | -   | -   | -  |
| Chitosan-Fe <sub>2</sub> O <sub>3</sub> -GO | 54                | 0.4 | 25.4 | 0.1 | 0.5 | 3.5 | 0.2 | 0.1 | 15 |

### 3.1.3. Characterization of Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composite using BET

The results of BET analysis on chitosan and chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite are shown in Table 2. Surface area determination using BET was performed on chitosan and chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite. The results are shown in Table 2. It shows that the surface area of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> is 39,946 m<sup>2</sup>/g, which is greater than the surface area of chitosan, which is 7.036 m<sup>2</sup>/g. This is due to the addition of iron oxide (Fe<sub>2</sub>O<sub>3</sub>), which can increase the porosity of the composite, and graphene oxide (GO), which also has a large surface area where the more comprehensive the surface of the adsorbent (the smaller the adsorbent size), the greater the ability to adsorb because the

substance attached to the surface of the adsorbent increases [38]. These findings confirm that the combination of Chitosan, Fe<sub>2</sub>O<sub>3</sub>, and GO synergistically increases the porosity, surface area, and number of active sites in the composite, making it very effective for pollutant adsorption applications. Supporting research, the role of the combination of iron oxides, graphene oxide (GO), and chitosan in increasing adsorption capacity has been reported in several studies. Research on composites comprising chitosan, graphene oxide, and Fe<sub>2</sub>O<sub>3</sub> conducted by previous researchers showed that these composites can be efficient and inexpensive adsorbents for removing dyes from aqueous solutions [39]. Other researchers had also reported that the Fe<sub>2</sub>O<sub>3</sub>-GO/CS composite was

environmentally friendly and could remove Cr(IV) ions with a removal efficiency of 94.3% [40].

**Table 2** Comparison of Surface Area of Chitosan and Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composite.

| Adsorbent                                  | Surface area (m <sup>2</sup> /g) |
|--------------------------------------------|----------------------------------|
| Chitosan                                   | 7.036                            |
| Chitosan/GO-Fe <sub>2</sub> O <sub>3</sub> | 39.946                           |

### 3.2. Methylene Blue Adsorption Using Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composite

#### 3.2.1. Determination of pH Point Zero Charge (pHpzc) on Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composite

pHpzc of the research conducted is presented in Fig. 6. In several studies, it was found that the adsorption of several substances is highly dependent on pH. The pzc value is determined by the adsorbent's characteristics. At lower pH, hydrogen ions (protons, H<sup>+</sup>) will be adsorbed more than other cations (adsorbates), so other cations will be adsorbed less if the particle is negatively charged. On the other hand, if the surface is positively charged and the pH increases, fewer anions will be adsorbed because hydroxide ions increase [41]. For adsorbents, if the pH is below the pzc, the surface charge is positive, allowing anions to adsorb. Conversely, if the pH is above the pzc value, the surface charge will be negative, so cations can be adsorbed. The results of the research that has been done on the

chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite show that there is an intersection point at the initial pH and final pH at pH 7.5, so it can be concluded that pHpzc is at pH 7.5 (Fig. 6). This study used methylene blue dye, where methylene blue here is cationic. The results of the study on the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite showed that the zero point of surface charge (pHpzc) was at pH 7.5, determined from the intersection of the initial and final pH curves [42]. This phenomenon indicates that at pH <7.5, the composite surface is positively charged, while at pH > 7.5 it is negatively charged. Although methylene blue is cationic (positively charged), the study showed that adsorption remains effective at pH conditions around pHpzc due to several supporting mechanisms, such as  $\pi$ - $\pi$  interactions between the aromatic structure of methylene blue and graphene oxide groups, hydrogen bonds between hydroxyl/amino groups on the composite with dye molecules and surface complexation through interactions with Fe<sub>2</sub>O<sub>3</sub> ions bound in the composite [37]. At the optimal pH of 9 (above pHpzc), the negative charge of the composite surface enhances adsorption through electrostatic attraction with methylene blue cations. These results are consistent with similar composite studies using chitosan-graphene oxide matrices for cationic dye adsorption [43].

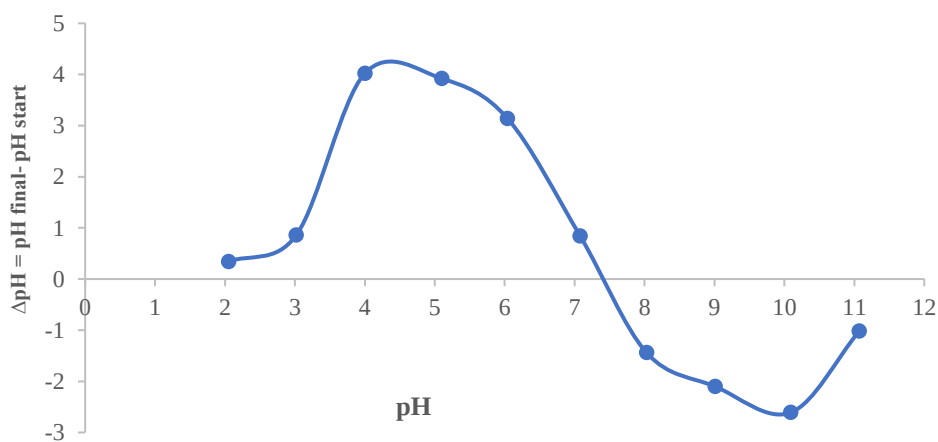


**Fig. 6** Determination of Point pHpzc of the Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> by the pH Drift Method.

#### 3.2.2. Effect of pH Variation on the Adsorption of Methylene Blue

The effect of pH variation on the methylene blue dye is shown in Fig. 7. The degree of acidity/pH can inhibit or encourage the adsorption process in solution. This is because the pH of the solution affects the ion form of the adsorbed dye, its charge, and the type of adsorbate on the adsorbent surface [44]. Figure 7 shows that the optimum pH for the absorption of methylene blue dye on the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite is at pH 9, with an absorption capacity of 7.29 mg/g. This indicates that at pH 9, there is no competition between H<sup>+</sup> ions and methylene blue [41]. At

pH above 9, the adsorption efficiency of methylene blue by chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composites decreased due to the competition between OH<sup>-</sup> ions and methylene blue molecules in occupying active sites on the adsorbent surface. Abundant OH<sup>-</sup> ions in basic conditions can compete with methylene blue to bind to the adsorbent surface, so that the number of dye molecules adsorbed decreases [37]. In addition, OH<sup>-</sup> ions can also form a protective layer around the adsorbent that inhibits the electrostatic interaction between positively charged methylene blue and the negatively charged adsorbent surface. This phenomenon has been observed, where

increasing pH above 9 significantly decreases the adsorption capacity of methylene blue due to competition with  $\text{OH}^-$  ions [45]. Other studies have also shown that the optimum pH condition for the adsorption of methylene blue by chitosan and graphene oxide-based composites is pH 9, with maximum adsorption capacity and high efficiency, but the performance decreases at higher pH due to the competition mechanism [46].

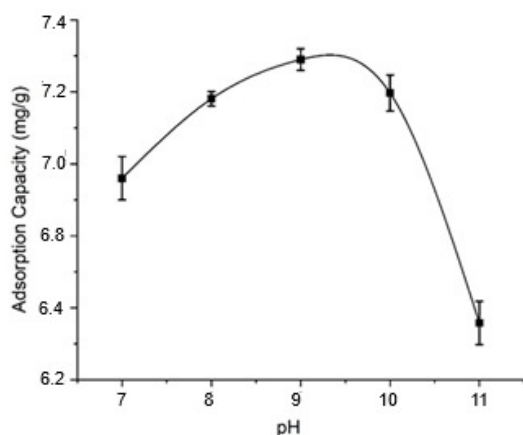


**Fig. 7** Effect of pH on the Adsorption Performance of the Adsorbent Chitosan/GO- $\text{Fe}_2\text{O}_3$ .

### 3.2.3. Effect of Contact Time Variation on the Adsorption of Methylene Blue

The effect of contact time variation on absorption capacity can be seen in Fig. 6. From the data produced at 10-50 minutes, there was an increase in absorption power because the longer the contact time, the greater the absorption power, but in the following minutes, there was a decrease in adsorption power due to the long time, so that the number of pores in the adsorbent experienced a saturated condition. This phenomenon is similar to the study by Ramadhani et al. [47].

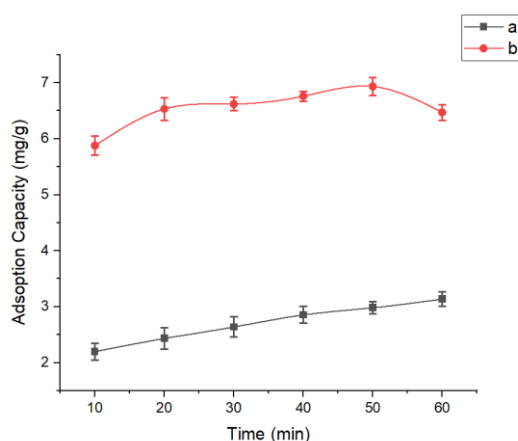


**Fig. 8** Effect of Contact Time Variation: (a) Chitosan and (b) Chitosan/GO- $\text{Fe}_2\text{O}_3$ .

The optimum contact time for methylene blue adsorption occurred at 50 minutes, with an absorption power of 6.934 mg/g and an adsorption of 92.454%. According to research by Dinh et al. [48], adsorption occurs through 2

stages, namely the first stage, where the dye molecules will spread to the surface when they have reached the boundary layer and then enter the pores of the adsorbent, and then the next stage, the pores of the adsorbent will experience saturation so that the adsorbed substance is released quickly. Generally, the adsorption rate increases, but over time it decreases to its equilibrium point. This is due to the large number of pores available for the adsorbate to enter, and this number will decrease over time. The pores left on the surface of the adsorbent will be challenging to occupy due to the repulsive force between the adsorbate molecules [49]. The adsorption capacity of chitosan adsorbed on methylene blue over time variation is smaller than that produced by the chitosan/GO- $\text{Fe}_2\text{O}_3$  composite. The adsorption capacity of the chitosan/GO- $\text{Fe}_2\text{O}_3$  composite as an adsorbent ranges from 6-7 mg/g, while the adsorption capacity of chitosan as an adsorbent is 2-3 mg/g. This is due to the composite's larger surface area compared to chitosan. With more pores, the composite can provide many active sites to adsorb the methylene blue dye.

### 3.2.4. Effect of Concentration Variation on the Adsorption of Methylene Blue

The effect of concentration variation on adsorption capacity is shown in Fig. 9. As observed, the optimum condition was achieved at a concentration of 40 mg/L, with an adsorption capacity of 11.29 mg/g and an adsorption efficiency of 94.081%. The adsorption capacity increased with increasing initial dye concentration, indicating that a higher concentration provides a stronger driving force to overcome the mass transfer resistance between the solid and liquid phases. However, when the adsorbent was exposed to a higher dye concentration of 45 mg/L, the adsorption capacity decreased. This reduction suggests that at higher concentrations, the available active sites on the adsorbent surface become saturated, leading to decreased adsorption performance or even partial desorption of the adsorbed molecules [50]. Chitosan adsorbed at various concentrations has a lower absorption capacity than that of the chitosan/GO- $\text{Fe}_2\text{O}_3$  composite. The adsorption capacity of chitosan/GO- $\text{Fe}_2\text{O}_3$  composite is 6-10 mg/g, while it is 2-3 mg/g for chitosan. Therefore, chitosan/GO- $\text{Fe}_2\text{O}_3$  composite is more effective for adsorbing methylene blue dye.

### 3.2.5. The Adsorption Isotherm Models of Methylene Blue

The adsorption isotherm describes the equilibrium between the adsorbent and the adsorbate. The Langmuir and Freundlich adsorption isotherm equations describe the adsorption process. The Langmuir isotherm assumes that adsorption occurs on a

homogeneous surface and that there is no interaction between adsorbates on the surface, whereas the Freundlich isotherm uses an

empirical equation that accounts for adsorption on heterogeneous surfaces [51]. Table 3 shows the data of the adsorption isotherm model.

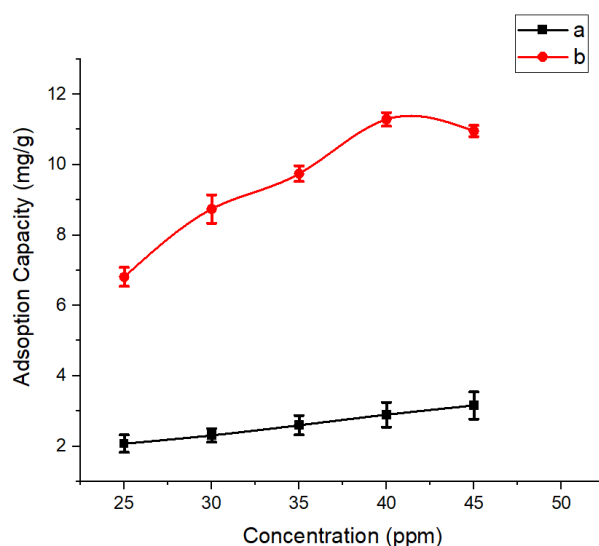


**Fig. 9** Effect of Concentration Variation: (a) Chitosan and (b) Composite.

**Table 3** Adsorption Isotherm Parameters of Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> Composites against Methylene Blue Dye.

| Isotherm Models | Parameter                       | Value  |
|-----------------|---------------------------------|--------|
| Langmuir        | $Q_m$ (mg/g)                    | 11.655 |
|                 | $K_L$ (L/mg)                    | 1.634  |
|                 | $R^2$                           | 0.9602 |
| Freundlich      | $N$                             | 9.2678 |
|                 | $K_f$ (mg/g)(mg/L) <sup>n</sup> | 8.468  |
|                 | $R^2$                           | 0.1812 |

The isotherm shape can convey information about the interaction between the adsorbate and the adsorbent and the molecules' adsorption affinity. The adsorption isotherm is based on a simplified physical explanation of adsorption, and others must correlate empirical data [52]. The table shows that the highest correlation coefficient ( $R^2$ ) value is in the Langmuir equation of 0.9602, while the Freundlich equation is 0.1812. Based on the correlation coefficient, the Langmuir correlation is close to 1, indicating that the isotherm follows the Langmuir adsorption

isotherm model. All active sites have equivalent adsorption energies. The  $Q_m$  and  $K_L$  values of the Langmuir isotherm are 11.655 (mg/g) and 1.634 (L/mg), respectively, where the adsorption capacity will be greater if the  $Q_m$  value is also significant [53]. The adsorption capacity of the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite produced was compared with several adsorbents, as presented in Table 4. It can be seen that the adsorbent used in this study is more effective than other adsorbents for adsorbing methylene blue dye.

**Table 4** Comparison of Adsorption Performance Between chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> and Other Adsorbents.

| Adsorbent                                             | Capacity adsorption (mg/g) | Author     |
|-------------------------------------------------------|----------------------------|------------|
| Chitosan/GO-Fe <sub>2</sub> O <sub>3</sub>            | 11.655                     | This Study |
| Zeolite-PBO composite                                 | 1.186                      | [54]       |
| Activated carbon (coconut shell) – Chitosan composite | 3.357                      | [55]       |
| Coconut Fiber Biosorbent                              | 4.467                      | [56]       |

### 3.2.6. The Kinetic Models of Methylene Blue Adsorption

The kinetic model of methylene blue dye adsorption using chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite based on the first-order and second-order kinetic models. The results of the adsorption kinetic determination are shown in Table 5. Based on Table 5, it can be seen that the  $R^2$  value of the pseudo-second order is higher than that of the pseudo-first order. This

indicates that the kinetic model for chitosan/Go- Fe<sub>2</sub>O<sub>3</sub> composite adsorption of methylene blue dye follows the pseudo-second order model. The  $q_e$  value in the pseudo-second order is 7.527 mg/g, which is not much different from the  $q_e$  value obtained from the experiment, which is 6.394 mg/g. The pseudo-second order kinetic model shows that the adsorption process may occur by chemisorption [57].

**Table 5** Kinetic Adsorption Profiles of Methylene Blue (MB) Chitosan/GO-Fe<sub>2</sub>O<sub>3</sub>.

| Kinetic Models      | Parameter                                              | Value  |
|---------------------|--------------------------------------------------------|--------|
| Pseudo First Order  | q <sub>e</sub> (mg/g)                                  | 1.873  |
|                     | k <sub>1</sub> (min <sup>-1</sup> )                    | 0.0637 |
|                     | R <sup>2</sup>                                         | 0.941  |
| Pseudo Second Order | q <sub>e</sub> (mg/g)                                  | 7.527  |
|                     | k <sub>2</sub> (g mg <sup>-1</sup> min <sup>-1</sup> ) | 0,0556 |
|                     | R <sup>2</sup>                                         | 0.999  |

#### 4.CONCLUSIONS

The synthesis of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite produces a blackish-green colour. The results of XRD characterization of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite show similarities between the 2θ peak and the 2θ peak in JCPDS data No. 33-0664 with angles of approximately 24.97°, 34.23°, 36.63°, 50.71°, and 55.17°. SEM-EDS analysis shows that the composite has a porous structure and contains C, N, O, and Fe elements. The BET characterization results obtained a surface area for the chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite of 39.946 m<sup>2</sup>/g. The ability of chitosan/GO-Fe<sub>2</sub>O<sub>3</sub> composite to adsorb methylene blue dye is best at pH 9, contact time of 50 minutes, and methylene blue dye concentration of 45 ppm with an adsorption capacity of 11.290 mg/g and an adsorption efficiency of 94.081%. The adsorption isotherm follows the Langmuir model, with an adsorption capacity of 11.655 mg/g.

#### ACKNOWLEDGEMENTS

"The research/publication of this article was funded by DIPA of the Public Service Agency of Universitas Sriwijaya 2021. Number SP DIPA 023.11.2.67751512024, On November 21, 2023. In accordance with the Rector's Decree Number. 0013/UN9/LP2M.PT/2024, On May 20, 2024".

#### NOMENCLATURE

|                      |                                                                           |
|----------------------|---------------------------------------------------------------------------|
| <i>d</i>             | Crystallite size, nm                                                      |
| <i>k</i>             | Scherrer constant, 0.96                                                   |
| <i>λ</i>             | X-ray wavelength, nm                                                      |
| <i>θ</i>             | Diffraction angle, °                                                      |
| <i>y</i>             | Absorbance                                                                |
| <i>x</i>             | Concentration                                                             |
| <i>a</i>             | Slope                                                                     |
| <i>b</i>             | Intersept                                                                 |
| <i>P</i>             | Percentage of adsorption, %                                               |
| <i>C<sub>0</sub></i> | Initial solution concentration methylene blue, mg/L                       |
| <i>C<sub>t</sub></i> | Solution concentration at time <i>t</i> , mg/L                            |
| <i>C<sub>e</sub></i> | Equilibrium concentration of adsorbate in solution after adsorption, mg/L |
| <i>Q</i>             | amount of adsorbate adsorbed per weight of adsorbent, mg/g                |
| <i>K<sub>L</sub></i> | Langmuir asorption constant, L/mg                                         |
| <i>b</i>             | Maximum adsorption capacity of the adsorbent, mg/g                        |
| <i>K<sub>f</sub></i> | Freundlich adsorption constant, L/g                                       |
| <i>Q<sub>e</sub></i> | Adsorption capacity at equilibrium, mg/g                                  |
| <i>V</i>             | Volume methylene blue, mg/L                                               |
| <i>W</i>             | weight of composite, mg                                                   |

#### Greek symbols

|          |                                        |
|----------|----------------------------------------|
| <i>β</i> | FWHM (Full Width at Half Maximum) peak |
|----------|----------------------------------------|

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